Temperature dependence of the spin dynamics in CuO layers

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The effect of temperature in the spin dynamics of the insulating CuO layers is investigated. The main thermally activated mechanisms are distinguished using data from infrared optical absorption experiments and calculating the spectra of multimagnon excitations assisted by phonons. Solving in finite systems a Heisenberg-like model, which includes a sizable four-spin cyclic exchange term, the high temperature spectra could be explained. At low temperatures the low energy absorption spectra is well described and the higher energy features are expected to be recovered in the thermodynamic limit. The processes found responsible for the main temperature dependence are: spin-phonon interaction and a shift into lower frequencies due to processes in which a thermally excited phonon is absorbed creating a bimagnon.

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The spin dynamics in the Cu-O planes has been subject of many different experimental and theoretical studies in the belief that spin degrees of freedom are important for the superconducting phase, being responsible for the pairing mechanism and phase-coherence. Therefore, it is important to understand how the spin degrees of freedom are affected by temperature (after all, the main challenge in the cuprates is to obtain a higher superconducting temperature). In this work it will be shown that spin-phonon coupling simulated by disorder plays an important role at finite temperature. Experimentally, this problem was first studied with Neutron and Raman scattering with inconclusive results. No model could explain the Raman B_{1g} "anomalous" features at low temperatures and most of the A_{1g} line. Resonant effects [1,2], are competing with mechanisms like spin-phonon interactions and four-magnon scattering, making the analysis of the Raman experiments very difficult. Recently, it has been shown that another class of experiments can provide information on the spin dynamics [3]. Lorenzana and Sawatzky developed a theory capable of explaining some peaks below the charge transfer gap in infrared optical absorption (IROA) experiments. Their theory is based on magnetic excitations assisted by phonons and is called Phonon Assisted Multimagnon Light Absorption (PAMLA). At $T = 0$, light is absorbed creating a phonon which lowers the symmetry of the electromagnetic problem and allows the excitation of a pair of magnons, otherwise forbidden. The pair of magnons evolves according to the magnetic Hamiltonian, contributing to the absorption of light. Therefore the spin response must be shifted in energy by the phonon's frequency ω_0. Their theory successfully explained the 2D S=1 La_2NiO_4 [3](b) and the 1D S=1/2 Sr_2CuO_3 [3](d) systems. In this way, the IROA experiments became a powerful tool to get information on the spin dynamics, without the problems involved in the Raman and Neutron experiments. Lorenzana and Sawatzky's theory applied to a Heisenberg model, could predict the features of the first narrow peak in the cuprates (T$\simeq$0), but could not explain the hight energy spectrum [4]. It has been shown that additional exchange interactions up to third nearest neighbors and four cyclic spin exchange terms account for the high energy part of IROA experiments in the thermodynamic limit. Moreover, this generalized Heisenberg model can reproduce the magnetic Raman and charge excitations in $Sr_2CuO_2Cl_2$ [5], and the B_{1g} and A_{1g} Raman responses in other seven cuprates [6] at room temperature.

The main goal of this work is to describe the principal effects of temperature in the spin dynamics of the cuprate layers, using the PAMLA theory at finite temperatures. The magnetic Hamiltonian is [4,6]:

$$H = \sum_{i,j} J_{ij} \boldsymbol{S}_i . \boldsymbol{S}_j + H_{4S} \tag{1}$$

$$H_{4S} = K \sum_{<i,j,k,l>} (\boldsymbol{S_i}.\boldsymbol{S_j})(\boldsymbol{S_k}.\boldsymbol{S_l}) + (\boldsymbol{S_i}.\boldsymbol{S_l})(\boldsymbol{S_j}.\boldsymbol{S_k}) - (\boldsymbol{S_i}.\boldsymbol{S_k})(\boldsymbol{S_j}.\boldsymbol{S_l})$$

$\boldsymbol{S}_i$ is a spin 1/2 operator. The first term has non zero elements J_{ij} only if i, j are up to third nearest neighbors. H_{4S} is the four cyclic exchange Hamiltonian and the sum over $< i, j, k, l >$ represents all four spins in a plaquete. This Hamiltonian includes all the interactions derived from an extended Hubbard model up to 4^{th} order in degenerate perturbation in the kinetic energy [6]. The parameters will be the same as in Refs. [4,5], which have been shown to be consistent with Raman, angular-resolved photoemission, neutron scattering and, most important, can recover the high energy features of the IROA experiments in the thermodynamic limit [4]. Once the right Hamiltonian is settled, the optical conductivity $\sigma(\omega)$ for a 2D system at $T \neq 0$ can be derived following Ref. [10]. Neglecting spin-phonon interactions and using Einstein phonons of frequency ω_0 as the phonons involved in the PAMLA mechanism (this

is known to be a good approximation for the CuO layers [3]), $\sigma(\omega)$ can be written as:

$$\sigma(\omega) = \sigma_0 \omega (1 - e^{-\beta\omega}) I(\omega)$$
$$I(\omega) = \sum_{\mathbf{q}} \lambda_{\mathbf{q}}^2 [(1+n_0) J_{\mathbf{q}}(\omega - \omega_0) + n_0 J_{\mathbf{q}}(\omega + \omega_0)] \quad (2)$$

where n_0 is the Bose occupation number for the phonon mode. σ_0 is a material dependent constant and

$$\lambda_{\mathbf{q}}^2 = \sin^2(\frac{q_x}{2})[\sin^2(\frac{q_x}{2}) + \sin^2(\frac{q_y}{2})].$$

Here it was assumed that the photon electric field is polarized in the x direction. The term $(1 - e^{-\beta\omega})$ can be approximated to 1 for the temperatures and frequencies of the problem. All the magnetic information is stored in the function,

$$J_{\mathbf{q}}(\omega) = \frac{1}{Z_{\rm mag}} \sum_{\mu\nu} e^{-\beta E_\nu} \left| \langle \mu | B_{\mathbf{q}}^x | \nu \rangle \right|^2 \delta(\omega + E_\nu - E_\mu) \quad (3)$$
$$B_{\boldsymbol{q}}^x = \frac{1}{\sqrt{N}} \sum_j e^{i\boldsymbol{q}.\boldsymbol{R}_j} \boldsymbol{S}_j . \boldsymbol{S}_{j+\hat{x}}$$

where $Z_{\rm mag} = Tr[e^{-\beta \hat{H}}]$, and N is the number of lattice sites.

For zero temperature, the second term in Eq.2 is zero (since $n_0 = 0$) and the sum over the thermally excited states ν should not be considered, recovering the expressions from Ref. [3]. For non zero the effects of temperatures can be classified as: a) A shift in ω_0 corresponding to processes in which a phonon thermally excited is destroyed by the incoming photon and, at the same time, creating two magnons (this processes correspond to the second term in Eq. 2); b) contributions to the Green function from thermally excited states (corresponding to the sum over ν in Eq. 3); c) increasing magnon-phonon interactions.

This work concentrates on a) and c), trying to determine how important these effects can be. The spin-phonon interaction is of most interest since it has been suggested that magnetoelastic effects can play a crucial role in the superconducting phase [11,12].

If a spin-phonon interaction is considered, the phonons can be simulated as Gaussian disorder introduced in the magnetic Hamiltonian. There have been several non-resonant approaches like this, trying to model the Raman response [6–8]. This Gaussian disorder can be thought as the result of an adiabatic approximation performed in the lattice distortions [8]. In fact, the characteristic Debye frequency is a third of the magnetic constant J [8]. But the first excited state lies around $3J$, so there is roughly an order of magnitude difference between magnetic and lattice excitations, justifying the adiabatic approximation. This effect is temperature dependent since the average number of phonons is temperature dependent. The spin-phonon interaction introduces magnon damping and the line will be broader as the temperature is increased [9]. A first indication that non-magnetic mechanisms, like spin-phonon interaction, are important is that the second moment of the Raman response is material-independent, although the exchange constant J for the cuprates can differ by 20% [6].

The calculation strategy followed in this work is: 1) the hopping matrix elements of a one band Hubbard model are given a certain Gaussian disorder, with a certain width ϵ, trying to mimic the effects of the phonons. 2) A strong coupling mapping from a Hubbard into an effective Heisenberg-like Hamiltonian (Eq.1) is performed. 3) The bimagnon Green's function in Eq.3 is calculated in clusters of 20 sites, using exact diagonalization [15]. 4) The Green function is averaged over 500 realizations of disorder. No quantum Monte-Carlo can be used since H has frustrating terms which lead to sign problem and no analytical solution or sensible approximation is known.

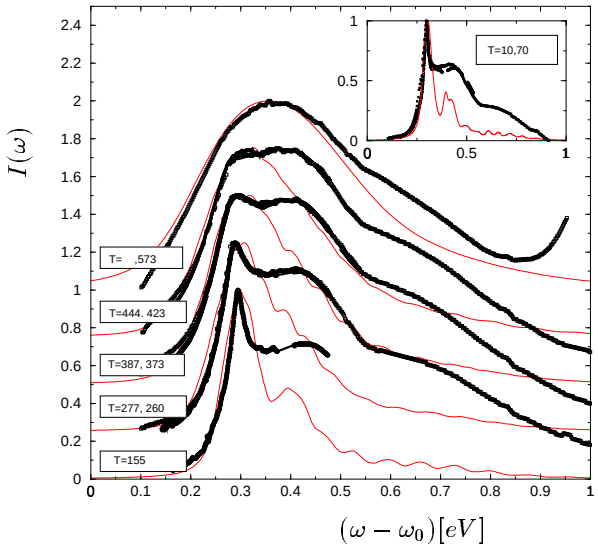

FIG. 1. $I(\omega)$ for different temperatures [in Kelvin] (see Eq. 2). The solid lines correspond to the numerical calculations including a spin-phonon coupling and a shift in ω_0 due to the second term in Eq.2. Most of the lines have two different curves corresponding to the data from [13] (first temperatures, solid circles) and from [14](second temperatures, empty squares) for $Sr_2CuO_2Cl_2$. In most of the cases they are completely indistinguishable. A linear background has been subtracted from the raw experimental data. The inset shows two different experimental results, showing the reproducibility and the similarity at low T, together with the non-disordered ($T = 0$) case. In the calculations $J = 0.134$ eV, and $\omega_0 = 0.06$ eV was used. The intensity scale was adjusted to match the first peak.

Fig. 1 shows the calculated $I(\omega)$ for different temperatures together with experimental data taken from Refs. [13,14]. Three different compounds ($Sr_2CuO_2Cl_2$, La_2CuO_4 and Nd_2CuO_4) have been studied in Ref. [13] and only $Sr_2CuO_2Cl_2$ in Ref. [14]. Both results for $Sr_2CuO_2Cl_2$ are in agreement. At low temperatures all compounds can be scaled into a single curve [4], indicating reproducibility and the fact that they are an inherent property of the CuO planes. In the following, for illustrative purposes only data for $Sr_2CuO_2Cl_2$ will be used. Typically, at low temperatures one can distinguish 3 peaks [13,14]. A narrow one around 0.3 eV, a broader one at 0.4 eV and a third one at 0.6 eV (see Fig. 1). The first two peaks can be associated to 2- and 4- magnon excitations in the Ising limit [3]. The main effect of H_{4S} and the second and third nearest neighbor Heisenberg like terms in the Hamiltonian (Eq. 1) is to induce mechanisms in which the 4 magnon processes are enhanced and therefore some spectral weight is transfered from the 2-magnon into the 4-magnon peak. As the temperature is increased, the first peak broadens and moves to lower frequencies, loosing weight with respect to the high frequency peaks.

In order to describe the Temperature effects in the IROA experiments, one needs to relate temperature and disorder. The disorder introduced by the phonons depends upon their displacement $< \delta x^2 >_{(T)}$:

$$\epsilon \sim \langle \delta x^2 \rangle_{(T)} = \frac{\hbar}{m\omega}(\langle n_0 \rangle_{(T)} + 1)$$

where $\langle n_0 \rangle_{(T)}$ is the Bose occupation number. For a particular phonon this function is linear for high Temperatures (higher than the phonon frequencies) Associating a disorder of $\epsilon = 22.5\%$ in the Hamiltonian to the 444K IROA spectrum, and knowing that in the Raman case at room-temperature a 13% disorder provides a good fit [6], the two points needed for the lineal relation are settled. At the same time, the Raman scattering disorder and the IROA experiments disorder (and therefore their temperature dependence through spin-phonon interactions) are related. All curves in Fig. 1 for $T \gtrsim 150K$ were determined using this linear relation. For $T \lesssim 150K$ the experimental $I(\omega)$ is almost T-independent and very similar to the $T = 0$ case. Consistently, no disorder was introduced (see inset in Fig. 1).

In the numerical calculations a value of $J = 0.134$ eV defines the energy scale and was chosen to adjust the $T \approx 0$ spectrum, and kept fixed for all other temperatures. In all cases a Lorentzian broadening of $0.08J$ was used for the poles obtained from the exact calculations. Since small clusters calculations cannot give the appropriate description of the high energy peaks (the correct weights being recovered only in the thermodynamic limit [4]) this work will be concentrated on the features of the first peak. Including both effects a) and c) the theoretical explanation seems quite satisfactory. At very high temperatures nearly all the spectrum corresponding to the first two peaks can be adjusted. In order to do the comparison more quantitative, Figs. 2 a) and b) display the position and width of the first peak as a function of temperature. To analyze the relevance of each mechanism, their separete contribution was considered: the spin-phonon coupling contribution is indicated with diamonds, while triangles show the combined effect of a) and c). The spin-phonon mechanism alone accounts for most of the width, but it alone cannot explain the position. The additional mechanism c) corrects for this deficiency.

The two effects a) and b) have different energy scales. The typical energy scale of a) corresponds to the energy of the optical phonon responsible for the PAMLA mechanism $\omega_0 = 0.06$ eV. The energy associated to the spin-phonon interaction is related to the acoustic phonon band. This acoustic band has frequencies as big as $\omega_1 = 0.04$ eV. The dependence in temperature of this two effects should be some function of the occupation number of this two different energy phonons. It is very instructive to fit the experimental data with the Bose occupation number corresponding to these two energies. The dashed line in Fig. 2 corresponds to a fit of the function $f_0(T) = 1 + A_0 n_0^{\omega_0}(T)$ and the solid to $f_1(T) = 1 + A_1 n_0^{\omega_1}(T)$, where A_0 and A_1 are the fitting parameters and $n_0^{\omega}(T)$ is the Bose function with energy ω. The Bose function with energy ω_1 can fit very well the low temperature data ($T \lesssim 300K$) in Fig. 2a) and the fit with ω_0 is very good for high temperatures ($T \gtrsim 400K$). The fact that $f_1(T)$ adjusts the low temperature data is in agreement with the numerical calculations taking into account *only* the spin-phonon interaction (the mechanism associated with ω_1), where the adjustment is reasonable for $T < 300K$ and then it is very poor. Only when the effect a) is also considered the agreement is good for high T. When trying this kind of fit to the width (Fig. 2b)) the low temperature data is very well adjusted ($T \lesssim 300K$) with $f_1(T)$. The same function also fits the numerical calculations up to $T \approx 450K$, indicating, once more, that only this mechanism is responsible for most of the width. $f_0(T)$ cannot satisfactory fit the width at no range of T.

The smaller width for the numerical calculations at high temperatures could be due to finite size effects. As the temperatures rises, the first two peaks become broader and superpose. The second one becomes even predominant [14]. As no good adjustment of the second peak in this clusters is possible, finite size effects coming from the second peak will be presumibly bigger for big T. The effect labeled as b) can account for the lost width also. It is interesting to note that even though the spectrum for high temperatures seems to be in very good agreement (see Fig. 1 top line) the width of the first peak is not well described (Fig. 2b)).

What is the main effect of disorder in the spin degrees of freedom? Disorder could be an efficient way to

simulate the damping of magnons in the spin Hamiltonian. As explained by Lorenzana and Sawatzky, at $T = 0$ the broadening of the first peak comes principally from the decay of a bimagnon into two free magnons. When the spin-phonon interaction through disorder is included an enhancement of this mechanisms occurs. First, the density of excited states broadens, giving new states into which the bimagnon could decay. In particular new states with lower energy for the free magnons will arise, shortening the lifetime of the bimagnon with $\boldsymbol{k} = (\pi, 0)$ (known to have very long lifetime and being the main responsible for the sharp first peak at $T = 0$ [3]). Second, the spatial symmetry selection rules no longer apply and new possible scattering processes can occur. This two effects combine in order to give a smaller lifetime of the bimagnons.

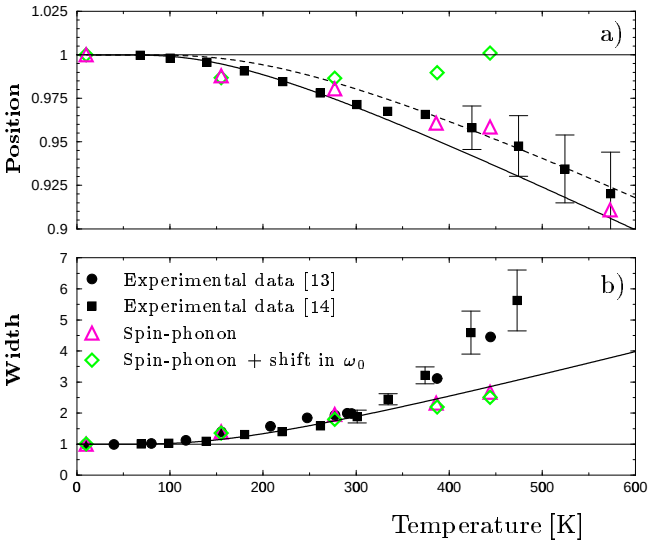

FIG. 2. a)Position and b)width for the first peak. All the values are relative to the lower temperature available. Diamonds correspond to the calculations taking into account only the spin-phonon coupling. Triangles include spin-phonon and a shift in ω_0. Solid lines are fits to the low temperature data with a function $f_1(T)$. The dashed line corresponds to a fit of the high temperature data with the function $f_0(T)$. Error bars are due to different possible fits of the Lorentzians to the data.

In conclusion, this paper addresses the importance of disorder induced by temperature on the spin dynamics of the CuO layers. In the IROA experiments two different processes are shown to be relevant: the shift into lower frequencies due to processes in which a thermally excited phonon is absorbed and creates a two magnon and spin-phonon interactions. Both mechanisms are important in order to explain the spectra at finite temperatures, accounting for different features at different temperatures. At high enough temperature, this processes explain most of the spectra for the first two peaks. At low temperatures, finite size effect become noticeable and the correct spectrum will be recovered in the thermodynamic limit. The good agreement between experiment and theory suggests once more that the main affect of spin-phonon interaction can be simulated as Gaussian disorder. It is interesting to note that Raman excitation and IROA experiments (where no resonant effects are expected) can be explain with the same values of disorder, strongly suggesting that the main "anomalous" effects in Raman scattering are non-resonant. It would be very useful to have experimental results for the temperature dependence of Raman scattering for the same compounds that IROA has been measured.

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[15] Notice that as all the translational symmetries are broken, this is the biggest size that can be solved and no reasonable scaling can be performed.